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The SiGe results were discussed in detail in the last AASERT report, so we
eschew them here. The F2 reactive scattering on Si(100) was the first study
in a series to ascertain the kinetics of surface processes, including etching
of silicon. We calculated reaction probabilities for F2 impinging on silicon,
as a function of translational and vibrational energy in the F2 molecules. We
find that translational excitation is slightly more effective than vibrational
excitation at increasing the reactivity of F2, but that vibrational excitation
is most effective for producing precursors leading to etching (SiF2). We find
that F-atom abstraction, where one Si-F bond is formed while the other F atom
in the F2 molecule leaves the surface, is a very probable reaction due to the
enormous exothermicity of the reaction (-100 kcal/mol exothermic do deposit only
one F on the silicon surface and -200 kcal/mol to deposit both F atoms). These
scattered F atoms do not have time to equilibrate with the surface, as is
illustrated by the fact that they are translationally hot and do not come off
in a cosine distribution. We have also recently studied the reaction of F2
molecules with stepped and defective Si(100) surfaces.

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AASERT Technical Report

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June 10, 1994

I have funding to support one student through the AASERT program. I have split this money between two students, Todd J. Martinez and Lawrence E. Carter. I now briefly describe their research progress. (Both of them have more than satisfactory grades.)

Todd Martinez is working on pseudospectral numerical approaches to the electron correlation problem, with the goal of developing accurate methods that scale better than current analytical methods such that larger molecules can be treated with configuration interaction (CI) or perturbation theory. In the past year, we have published one paper and one book chapter is due to appear this summer (reprints/preprints of which were sent to AFOSR already): the paper is on pseudospectral many-body perturbation theory through third order [*Journal of Chemical Physics*, 100, 3631 (1994)] and the book chapter is a pedagogical review of pseudospectral electron correlation methods emphasizing its relationship to other approaches (to appear in "Modern Electronic Structure Theory," edited by D. R. Yarkony, in the World Scientific Advanced Series in Physical Chemistry). We have shown that pseudospectral many-body perturbation theory calculations in fact show a distinct advantage in scaling while retaining chemical accuracy. Indeed, we have demonstrated that our pseudospectral MP2 code is an order of magnitude faster than existing commercial codes (Gaussian 92) for moderate-sized molecules with modest basis sets, and is expected to be even faster for molecules with extensive basis sets. Our pseudospectral MP3 code is extremely accurate and also faster than Gaussian 92, with expected speedups of more than an order of magnitude for large molecules. Todd is just finishing his final thesis project, developing pseudospectral multireference single and double excitation configuration interaction for arbitrary spin states, and that manuscript is in preparation. This method turns out to be even more accurate than our MP2 method (average error for a series of molecules is 0.2 kcal/mol in the total energy), and faster than conventional spectral methods. We are still optimizing the code, so final speedups relative to conventional methods are still to come. We are extremely encouraged by the speed and accuracy of these new methods for treating electron correlation that we have developed in the last few years, and expect to continue our development of these methods in the years to come.

Lawrence E. Carter has carried out Monte Carlo and ab initio-derived molecular dynamics simulations of SiGe superlattice structures and F_2 and F atom/Si(100) reaction dynamics. Two papers have been published in the last year, with another in press (these latter three have been sent to the AFOSR in preprint and reprint form) and several more in preparation [*Journal of Vacuum Science and Technology A*, 11, 2059 (1993); *Journal of Chemical Physics*, 100, 2277 (1994); *Journal of Vacuum Science and Technology A, July/August issue, 1994*]. The SiGe results were discussed in detail in the last AASERT report, so we eschew them here. The F_2 reactive scattering on Si(100) was the first study in a series to ascertain the kinetics of surface processes, including etching of silicon. We calculated reaction probabilities for F_2 impinging on silicon, as a function of translational and vibrational energy in the F_2 molecules. We find that translational excitation is slightly more effective than vibrational excitation at increasing the reactivity of F_2, but that vibrational excitation is most effective for producing precursors leading to etching (SiF_2). We find that F-atom abstraction, where one Si-F bond is formed while the other F atom in the F_2 molecule leaves the surface, is a very probable reaction due to the enormous exothermicity of the reaction (>100 kcal/mol exothermic to deposit only one F on the silicon surface and >200 kcal/mol to deposit both F atoms). These scattered F atoms do not have time to equilibrate with the surface, as

is illustrated by the fact that they are translationally hot and do not come off in a cosine distribution. We have also recently studied the reaction of F_2 molecules with stepped and defective Si(100) surfaces. We find that because the reaction is so exothermic, the reaction is insensitive to the presence of steps or defects, and indeed is merely concerned with the number of dangling bonds (sp^3 hybrid radical electrons) available to form bonds to F. This an extremely useful result from the experimentalist's perspective, because it says that it does not matter how defective your surface is, it should have similar reactivity towards F_2. We have also carried out extensive work on the reactive scattering of F_2 and F atoms on partially fluorinated surfaces and at higher incident translational energies of the F_2 molecules. We have discovered we can tailor the branching ratio between two reaction pathways just by varying the translation energy of the F_2, with atom abstraction dominant at low energies and dissociative chemisorption dominant at high energies. Lots of other interesting dynamical trends are emerging, that lend themselves to designing a new means of anisotropic etching of Si. We will send preprints as they are written up this summer, with more details, since I understand this report was to be only a very brief summary of the work.